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REBURNING THERMAL AND CHEMICAL PROCESSES IN A TWO-DIMENSIONAL PILOT-SCALE SYSTEM

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ABSTRACT

An experimental investigation of the thermal and chemical processes influencing NO_x reduction by natural gas reburning in a two-dimensional pilot-scale combustion system is described. Reburning effectiveness for initial NO_x levels ranging from 50 to 500 ppm and reburn stoichiometric ratios ranging from 0.75 to 1.0 is evaluated in the low temperature environment (less than 1500K) and simple aerodynamics of a firetube package boiler. Both input/output and in-flame sampling trials are reported to provide insight into the controlling kinetic mechanisms. An overall reaction order with respect to initial NO between 1.5 and 1.6 was observed. N_2 formation is observed to occur in both the fuel-rich reburning and fuel-lean burnout zones. NO destruction in the reburning zone is attributed to the reaction $CH + NO \longrightarrow HCN + O$, with less than 1 ppm CH radical required to produce the observed trends in nitrogenous species. Burnout zone N_2 formation is influenced by total fixed nitrogen speciation with low fractional conversion of HCN and NH_3. Under low primary NO conditions the reaction $CH + N_2 \longrightarrow HCN + N$ becomes important and is a contributor to reduced reburning effectiveness.

INTRODUCTION

Experimental results have been available for at least 30 years indicating that NO_x could be destroyed by reaction with hydrocarbons.[1,2] Fundamental study of NO_x destruction by injection of secondary fuel into primary combustion gases was performed by Wendt et al.[3], who coined the name "reburning." Concerted effort to develop reburning for application to large boilers occurred in Japan during the late 1970's and early 1980's. Takahashi et al.[4] document an extensive reburning development effort, and refer to the technique as Mitsubishi Advanced Combustion Technology, or MACT. They report that MACT applied to full scale boilers results in 50 percent NO_x reduction, independent of the fuel type and initial NO_x level. The current study reports on pilot-scale reburning experiments conducted in the lower temperature regime characteristic of small commercial firetube package boilers. The experiments were designed to provide insight into the controlling kinetic mechanisms of the reburning process.

EXPERIMENT

A complete description of the experimental system and the analytical techniques used in this study is available elsewhere.[5] Only a brief summary is presented here.

Apparatus

The combustion facility employed, shown schematically in Fig. 1, consists of a firetube package boiler simulator fitted with a low-NO_x burner. The boiler simulator is rated at 0.88 MW heat

input, is 0.6 m in diameter, and is 3.0 m long. The precombustion chamber burner provides control over the primary zone NO concentration and produces nearly constant radial species concentration profiles in the postflame gases.

Natural gas reburning fuel and burnout air were injected at the 1.02 and 1.62 m axial locations, respectively, using water-cooled concentric booms inserted through a port at the rear of the furnace. The inner boom, providing for secondary fuel injection, was equipped with 8 radial spokes, each with 12 fuel outlet ports oriented circumferentially. This configuration provided for rapid mixing between the secondary fuel and primary zone effluent. The location of the ports in each spoke was established such that the secondary fuel flow matched the flux of free oxygen from the primary zone, thereby providing radial uniformity in the reburn zone stoichiometry.

Analytical Methods

Stack and in-flame gas temperature, velocity, and speciation were measured. Probes were inserted through the boiler wall to determine radial characteristics at the reburn zone entrance, while axial variations were examined by inserting probes through ports in the boiler end plate. Flame temperature was measured using suction pyrometry. Velocities were measured using a Prandtl probe. Continuous monitors were used to determine CO, CO_2, UHC, O_2 and NO concentrations. Flame zone HCN and NH_3 concentrations were determined using specific ion electrodes. A water-cooled, water-quench, stainless steel sampling probe was used to extract in-flame gas samples.

Approach

All reported tests were performed with the boiler fired on natural gas at a constant primary load of 0.59 MW and a nominal primary zone excess air level of 7 percent. The primary zone NO_x level (NO_{pri}) was varied by doping the primary burner fuel with a trace quantity of ammonia, while reburn zone stoichiometry (SR_r) was varied by the secondary fuel injection rate. Furnace exit excess air was held constant by adjusting the final air addition rate.

The experimental program was divided into two components. First, baseline operation and overall reburning effectiveness for variations in SR_r and NO_{pri} were evaluated in a series of input/output trials. Second, the reburn zone chemical and thermal environment was evaluated in detail. Results from the detailed sampling trials were then compared to the input/output trial results to shed insight into the controlling chemical mechanisms.

RESULTS

Test results from the extensive series of input/output trials were reported previously[5,6] and are only summarized here to provide a basis for evaluating the in-flame results.

Input/Output Trials

A series of input/output tests were performed in which NO_{pri} was varied between 50 and 500 ppm and the value of SR_r varied between 0.75 and 1.0. Results are presented in Fig. 2 as lines of constant NO_{pri} versus SR_r. As shown, the extent of NO reduction is a function of both NO_{pri} and SR_r. The strong dependence on

NO_{pri} is in agreement with the reburning results reported by Chen et al.[7] Both the current results and those of Chen are in conflict with the MACT claim[4] that fractional NO destruction is independent of NO_{pri}. Analysis of the data in Fig. 2 reveals that the overall NO destruction rate has a partial order with respect to NO_{pri} between 1.5 and 1.6.

Temperature, Velocity, and Major Species Profiles

To better understand the influence of NO_{pri} and SR_r on the reburning process, a series of detailed flame probing experiments were conducted. Radial traverses were made to determine the temperature, axial velocity and major species concentration profiles at the entrance to the reburn zone. Temperature and velocity profiles are presented in Fig. 3. The velocity profile reaches a peak level of approximately 13 m/sec and has the expected shape for a non-swirling flow with a boom located on the centerline. Peak gas temperature of approximately 1400K occurs near the centerline boom and decreases to approximately 1150K at the furnace wall.

Samples of a tracer gas extracted from three radial locations just downstream of secondary fuel injection indicate that the secondary fuel injector provided both rapid mixing and radially uniform stoichiometry in the reburn zone. Based on this result a nominal probe position 13.3 cm from the centerline was selected for characterizing the fuel-rich chemistry. The axial velocity at this probing location is 8 m/sec. With the reburn zone length set at 0.6 m, the reburn zone residence time at this location is approximately 75 msec, while the residence time at the peak velocity radial location is approximately 50 msec. Parametric

variation of the final air injection location indicated that maximum reduction of the exhaust NO occurred with a reburn zone length of 0.6 m. This indicates that the characteristic time for processing nitrogenous species in the fuel-rich reburn zone is on the order of 50 msec.

As regards thermal environment, the extent of temperature increase from secondary fuel injection should be limited. After free oxygen from the primary zone is consumed, further oxidation of secondary fuel proceeds according to the overall reaction

$$CO_2 + CH_4 \longrightarrow 2CO + 2H_2 \qquad (1)$$

which is endothermic by 71.8 kcal/mole. Figure 4 presents measured axial profiles of reburn zone temperature for SR_r=0.95, 0.86 and 0.78. Also noted on Fig. 4 is the calculated adiabatic flame temperature for each of the test conditions. For SR_r=0.95 the measured temperature is slightly above the adiabatic flame temperature. For higher fuel injection rates (SR_r=0.86 and 0.78) the measurements indicate a slight temperature increase, while the corresponding adiabatic flame temperatures are lower than the primary zone exit temperature. The existence of super-equilibrium flame temperatures suggests less than complete decomposition of the injected secondary fuel. This is confirmed in Fig. 5 which presents the measured UHC concentration profile (expressed as methane) for the SR_r=0.86 case.

Nitrogen Chemistry

The primary thrust of the current study is to better understand the processing of nitrogenous species in the fuel-rich reburn zone. The effect of SR_r was examined in a series of trials where reburn zone stoichiometry was varied while holding NO_{pri}

constant. The influence of NO_{pri} was examined in a series of trials holding SR_r constant and varying NO_{pri}.

SR_r Variation Trials: Figure 6 presents fixed nitrogen species concentration profiles for constant NO_{pri} (~200 ppm) at SR_r values of 0.97, 0.86 and 0.78. The total fixed nitrogen (TFN) curve represents the summation of NO, HCN, and NH_3. A change in TFN reflects a change in the molecular nitrogen concentration. The indicated values of NO (and TFN) at the entrance to the reburn zone represent measured NO at the primary zone exit corrected for dilution by the secondary fuel. Also shown on Fig. 6 is the measured exhaust NO_x level and the exhaust NO level which would have existed if 100 percent of the reburn zone TFN had been converted to NO. The 100 percent TFN conversion levels are corrected for the molar change by burnout air addition and are expressed on a dry basis corrected to zero percent O_2.

The time constant for TFN decay (or N_2 formation) is on the order of 10 to 20 msec. Between 20 and 50 msec the NO concentration continues to decay but this destruction is accounted for by formation of HCN and NH_3. Thus, after a brief period of N_2 formation, the reburn zone serves to redistribute the TFN speciation.

As indicated by the difference between the 100 percent TFN conversion level and the exhaust NO_x level, significant N_2 formation occurs when burnout air is added. The extent of N_2 formation in this region is a fairly strong function of SR_r, with more N_2 being formed as SR_r is reduced. As the reburn zone becomes more fuel-rich a larger percentage of the TFN is in the form of HCN and NH_3. This indicates that the fractional conversion of HCN and NH_3 to NO is less than the fractional retention of NO.

Primary NO Variation: Figure 7 presents reburn zone nitrogenous species concentration data at SR_r=0.86 for three NO_{pri} levels: 206, 122 and 49 ppm. The characteristic time for the fixed nitrogen species reaction is again seen to be on the order of 50 msec. For all values of NO_{pri} the NO concentration is reduced by about 45 percent. The TFN profiles in Fig. 7 clearly reflect the decrease in overall NO_x reduction with decreasing NO_{pri} seen in the input/output trials. For the 122 and 49 ppm NO_{pri} trials, the TFN profiles indicate net destruction of N_2 in the fuel-rich reburn zone. The data in Figs. 6 and 7 show that reburning is influenced by two competing mechanisms, one which forms N_2 and one which destroys N_2. At high NO_{pri} levels the N_2 formation mechanism dominates while at low NO_{pri} the N_2 destruction mechanism becomes increasingly important.

It should be noted that, even though there was a net destruction of N_2 in the reburn zone for the low NO_{pri} trials, there was a net overall reduction of exhaust NO_x by reburning. For these low NO_{pri} tests overall NO_x destruction may be attributed to partial conversion of NH_3 and HCN to NO in the final air addition region of the process.

EVALUATION OF DOMINANT KINETIC PATHS

Chen et al.[7] have suggested that the path for NO destruction in the fuel-rich region is initiated by the reaction

$$CH + NO \longrightarrow HCN + O. \qquad (2)$$

The HCN participates in shuffle reactions leading to the formation of a partially equilibrated pool of NH_i species. The amine

radicals either react with NO to produce N_2 or are oxidized to reform NO. It is hypothesized here that the mechanism proposed by Chen dominates under high NO_{pri} conditions, and that an N_2 destruction process is important at low primary NO levels. Specifically, it is proposed that the N_2 destruction process is the first step in the "prompt NO" mechanism

$$CH + N_2 \longrightarrow HCN + N. \qquad (3)$$

NO Destruction Mechanism

The data in Fig. 6 indicate that reburn zone N_2 formation occurs for a limited time (~20 msec) and that conversion of NO to HCN continues for approximately 50 msec. Termination of N_2 formation is characterized by complete consumption of available free oxygen and the end of rapid hydrocarbon decomposition. This suggests that under low temperature conditions the O-H radical pool concentration is not high enough to sustain rapid burning and that the lack of O, H and OH radicals limits N_2 formation. The mechanism suggested by Chen requires OH and H radicals for conversion of HCN to NH and for the processing of NH to either N_2 or NO. The fact that the HCN concentration is significantly greater than the NH_3 further supports the contention that N_2 processing is limited by the O-H radical pool concentration.

In the early part of the fuel-rich reburn zone, where the radical pool concentration is likely to be high, N_2 formation could occur by either the mechanism suggested by Chen or a simple reverse Zeldovich mechanism. The current data set does not provide sufficient detail to identify the dominant mechanism. Beyond 20 msec there is substantial destruction of NO, producing HCN and a lesser quantity of NH_3. In this region, reaction 2 is a

plausible explanation for the observed trend. A basic question, however, is whether the CH radical concentration is sufficient to account for the observed NO processing. Direct measurement of the CH radical was not attempted, but it is possible to estimate the CH concentration required to produce the observed trends. To make that estimate it is assumed that HCN is being formed by reaction 2 and that any destruction of HCN results in formation of NH_3. This assumption must be restricted to high NO_{pri} conditions where N_2 destruction reactions are of minimal significance. In the region of constant TFN the experimental data in Fig. 6 may be used to estimate the concentration of the CH radical according to

$$Y_{CH} = \frac{U}{C_T k_{2,f} Y_{NO}} \frac{d(Y_{HCN} + Y_{NH3})}{dX} \qquad (4)$$

where U is the gas velocity and C_T is the molar gas density. The forward rate constant for reaction 2 is taken as $1x10^{13}$ cm^3/mole.s according to the estimate by Shaub and Bauer.[8] The calculated CH concentration profile, presented in Fig. 8, shows that less than 1 ppm of CH is sufficient to produce the observed variation in NO, HCN and NH_3. Considering the level of unburned methane in the fuel-rich reburn zone it is certainly plausible that several hundred ppb of CH could be present and that reaction 2 could be responsible for the reburn zone destruction of NO.

Molecular Nitrogen Destruction Mechanism

Figure 7 shows that reburning at low NO_{pri} is influenced by an N_2 destruction mechanism. It has been proposed that reaction 3 is responsible. To evaluate this hypothesis the experimental data are used to calculate a rate constant for reaction 3 and to compare the calculated rate to literature values. It is

recognized that the pilot-scale facility used in these experiments is not sufficiently controlled for development of reliable kinetic rate data. Rather, the intent is to establish whether the hypothesized mechanism is a reasonable explanation for the observed phenomena.

By assuming that HCN is formed by both reactions 2 and 3, that NH_3 is produced from HCN, and that N_2 production from reaction with NH_i is negligible, the forward rate of reaction 3 may be expressed as

$$k_{3,f} = \frac{U}{C_T Y_{CH} Y_{N_2}} \frac{d\left(Y_{HCN} + Y_{NH3}\right)}{dX} - k_{2,f} \frac{Y_{NO}}{Y_{N_2}} \quad . \qquad (5)$$

To apply Eq. (5) it is necessary to estimate the concentration profile of CH. In discussing the results presented in Fig. 7 it was noted that for constant SR_r the normalized NO profiles in the reburn zone exhibited first order behavior. Assuming that NO reduction is controlled by reaction 2, this first order NO behavior indicates that the CH radical concentration is dominated by hydrocarbon oxidation reactions and that reaction with nitrogenous species does not represent a major CH sink. Thus, the CH profile presented in Fig. 8 provides the approximate CH radical concentration for low NO_{pri} conditions. Figure 9 presents the values of $k_{3,f}$ calculated from Eq. (5) for NO_{pri} = 47 ppm. Also shown in Fig. 9 are the rates for reaction 3 calculated from the expression derived by Blauwens *et al.*[9] and from the rate estimate developed by Benson *et al.*[10] As seen, the reaction rate estimated by Eq. (5) falls between the two literature estimates. This agreement lends substantial credence to the proposed kinetic rationalization of the current reburning data.

CONCLUSIONS

The foregoing experimental investigation provides insight into the thermal and chemical processes associated with application of natural gas reburning to firetube package boilers. The extent of overall NO_x reduction is dependent on both NO_{pri} and SR_r. The rate of overall NO_x reduction was found to have a partial order with respect to NO_{pri} between 1.5 and 1.6.

With the boiler primary zone operated at low excess air levels, secondary fuel injection has a limited influence on thermal environment due to the endothermic nature of the overall fuel decomposition reaction. In the current application of reburning to a low temperature primary flame, the result was incomplete fuel decomposition in the fuel-rich region.

Under high NO_{pri} conditions N_2 formation was limited to the first 20 msec of the reburn zone. It was not possible to isolate the mechanism responsible for this N_2 formation. Between 20 and 50 msec substantial conversion of NO to HCN was observed. This conversion is attributed to reaction 3, requiring less than 1 ppm of CH radical to produce the observed trends. Only minor conversion of HCN to NH_3 occurred. Failure to produce amines or additional N_2 in the reburn zone appears to be the result of a depleted O-H radical pool concentration. Additional N_2 formation occurred when final burnout air was added. The data indicate that fractional conversion of HCN and NH_3 to NO is less than the retention of NO.

Under low NO_{pri} conditions destruction of N_2 by reaction with CH radicals is indicated. The experimental data are consistent

with literature values for the rate of reaction 3. This N_2 destruction process contributes to reduced NO_x control by reburning at low NO_{pri}. The characteristic time for reburn zone processing of nitrogeneous species was found to be approximately 50 msec for all experimental conditions examined. This characteristic time is consistent with results from input/output tests.

The conclusions drawn from these experiments are restricted to the low temperature environment of firetube package boilers and to reburning with natural gas. The overall consistency of the pilot-scale results with fundamental considerations indicates that the findings are not hardware specific.

ACKNOWLEDGMENTS

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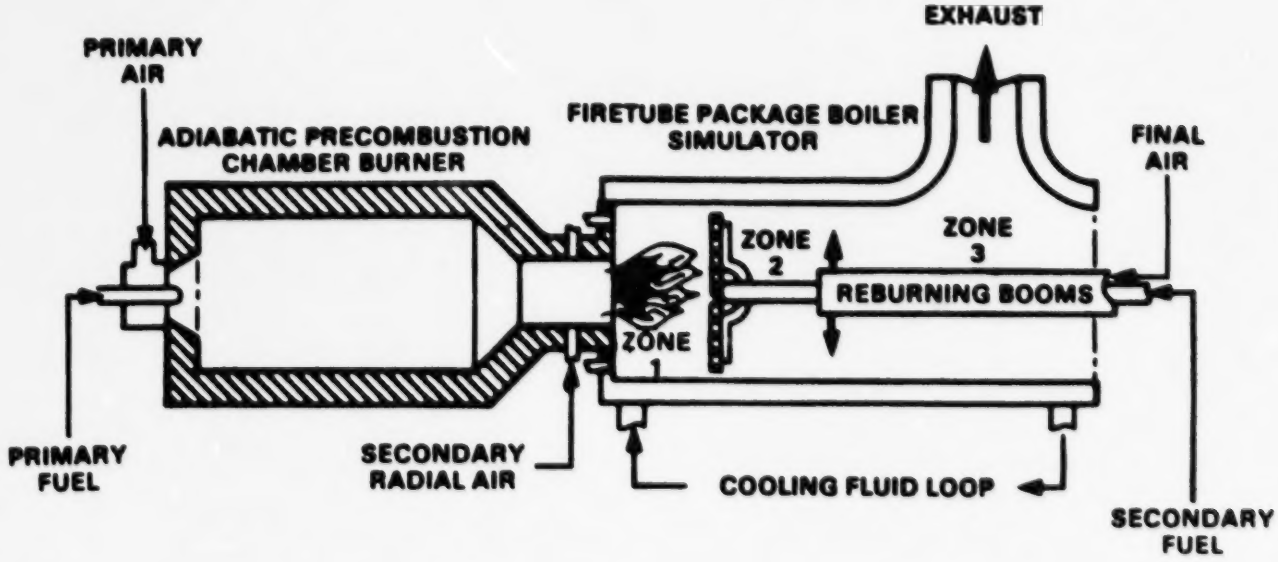

Figure 1. Pilot-scale combustion facility schematic, shown with reburning application. Zone 1 is the primary flame zone; zone 2 is the fuel-rich reburn zone; and zone 3 is the burnout zone.

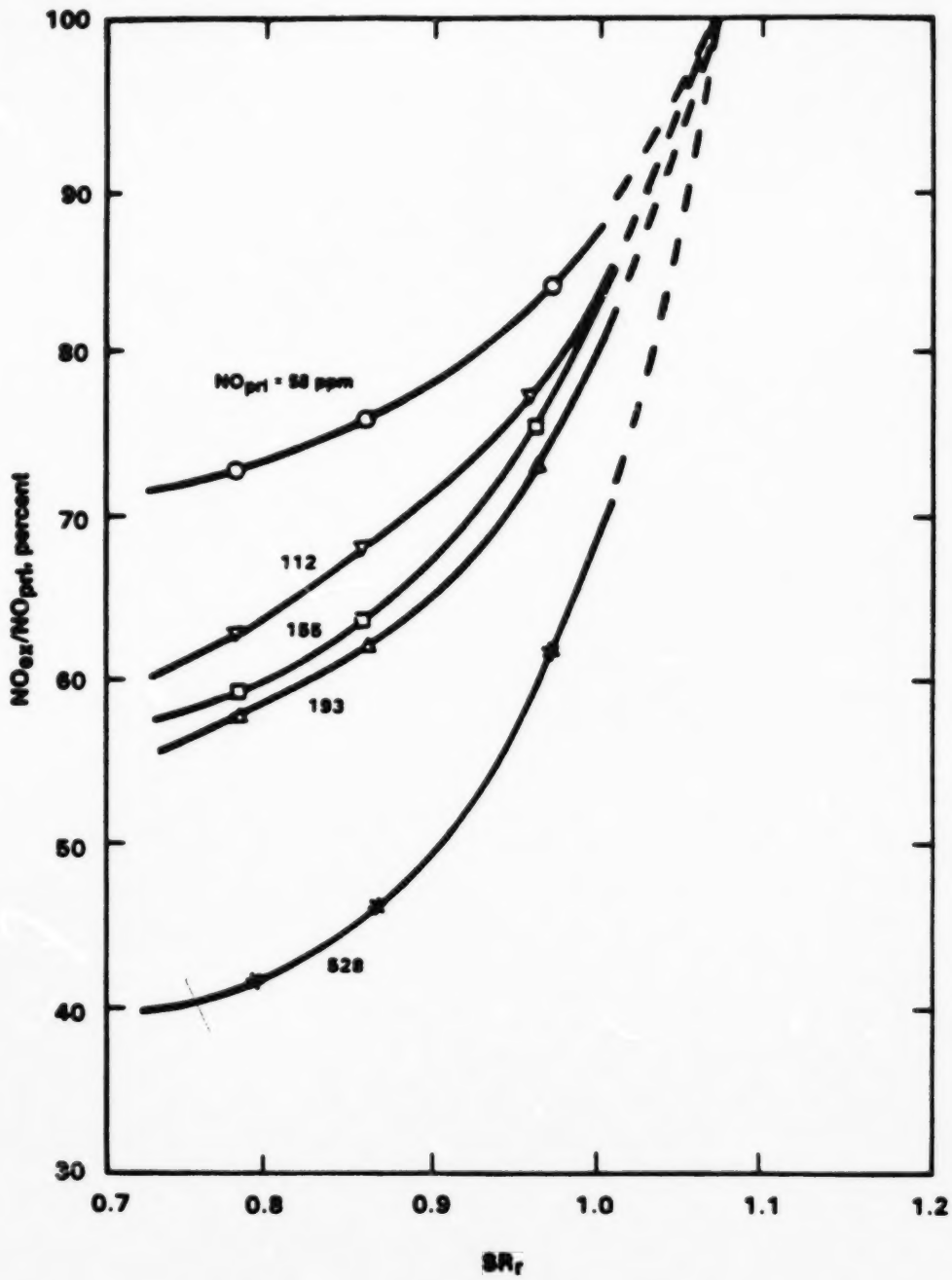

Figure 2. Overall reburning effectiveness with natural gas. Exhaust NO, plotted as a percent of primary NO, is shown as a function of reburn zone stoichiometry. Data are plotted for five levels of primary NO at three reburn zone stoichiometries.

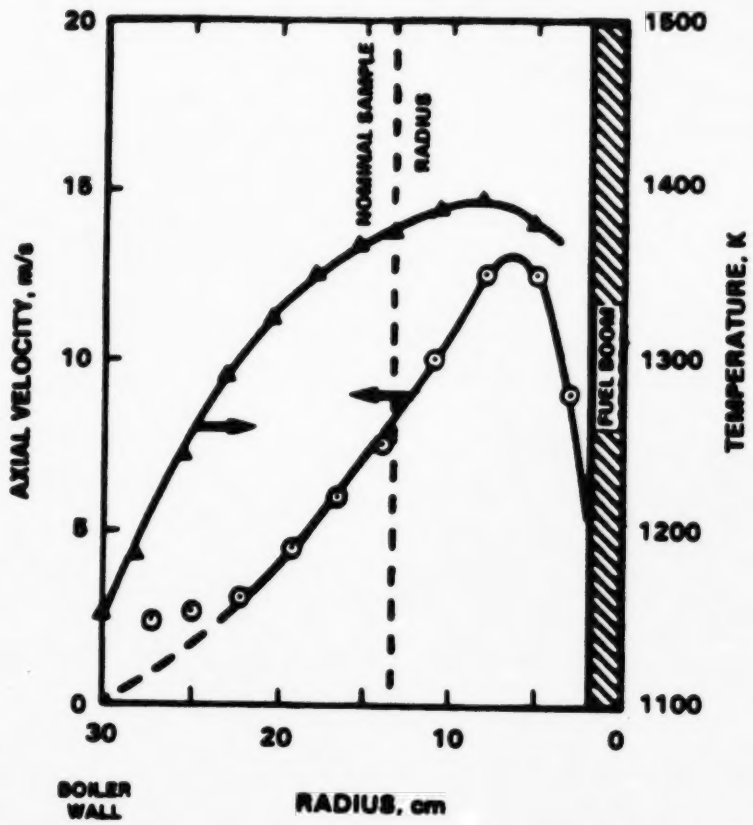

Figure 3. Temperature and velocity profiles entering the reburn zone.

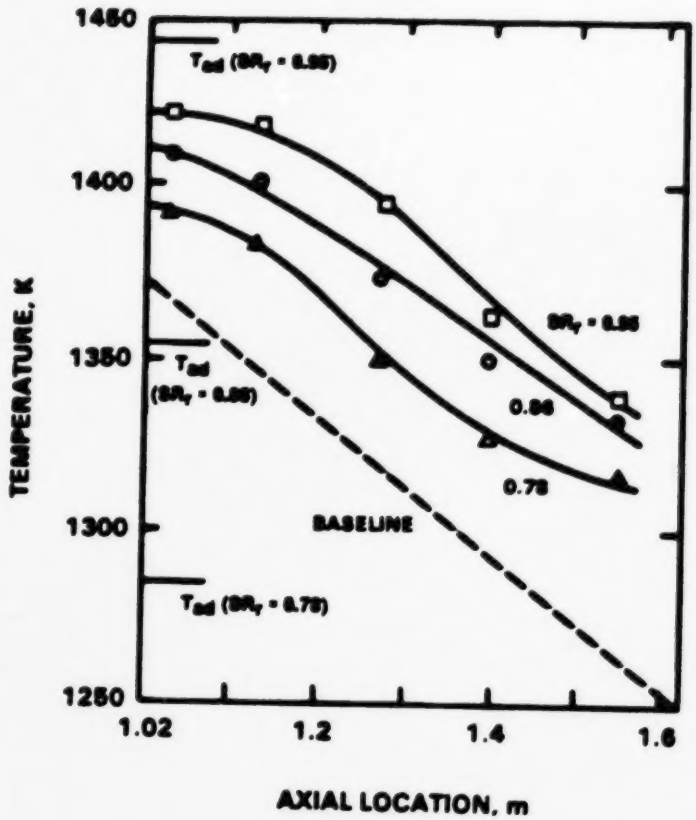

Figure 4. Axial temperature profiles in the reburn zone for three reburn load conditions.

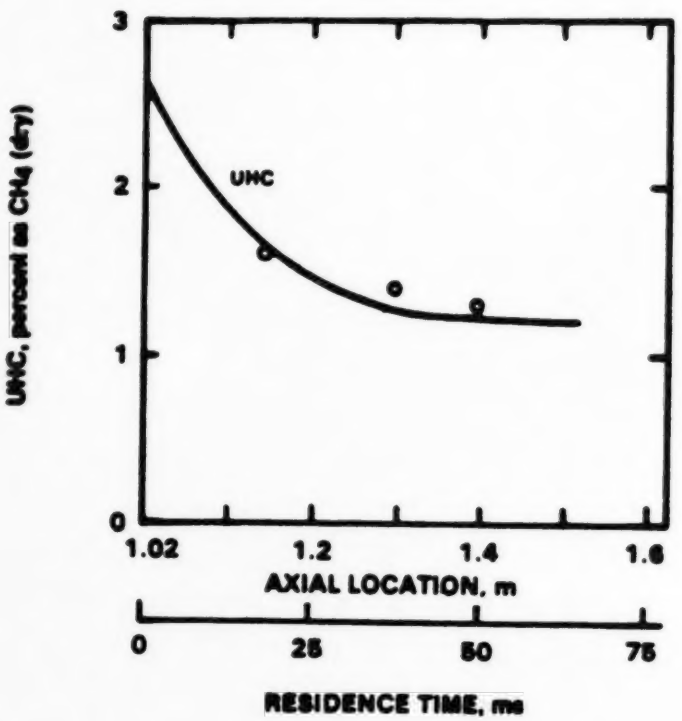

Figure 5. Unburned hydrocarbon profile for SR_r=0.86. The line represents a calculated CH_4 profile based on carbon atom balance and measurement of CO_2 and CO profiles. The data points represent actual measurements.

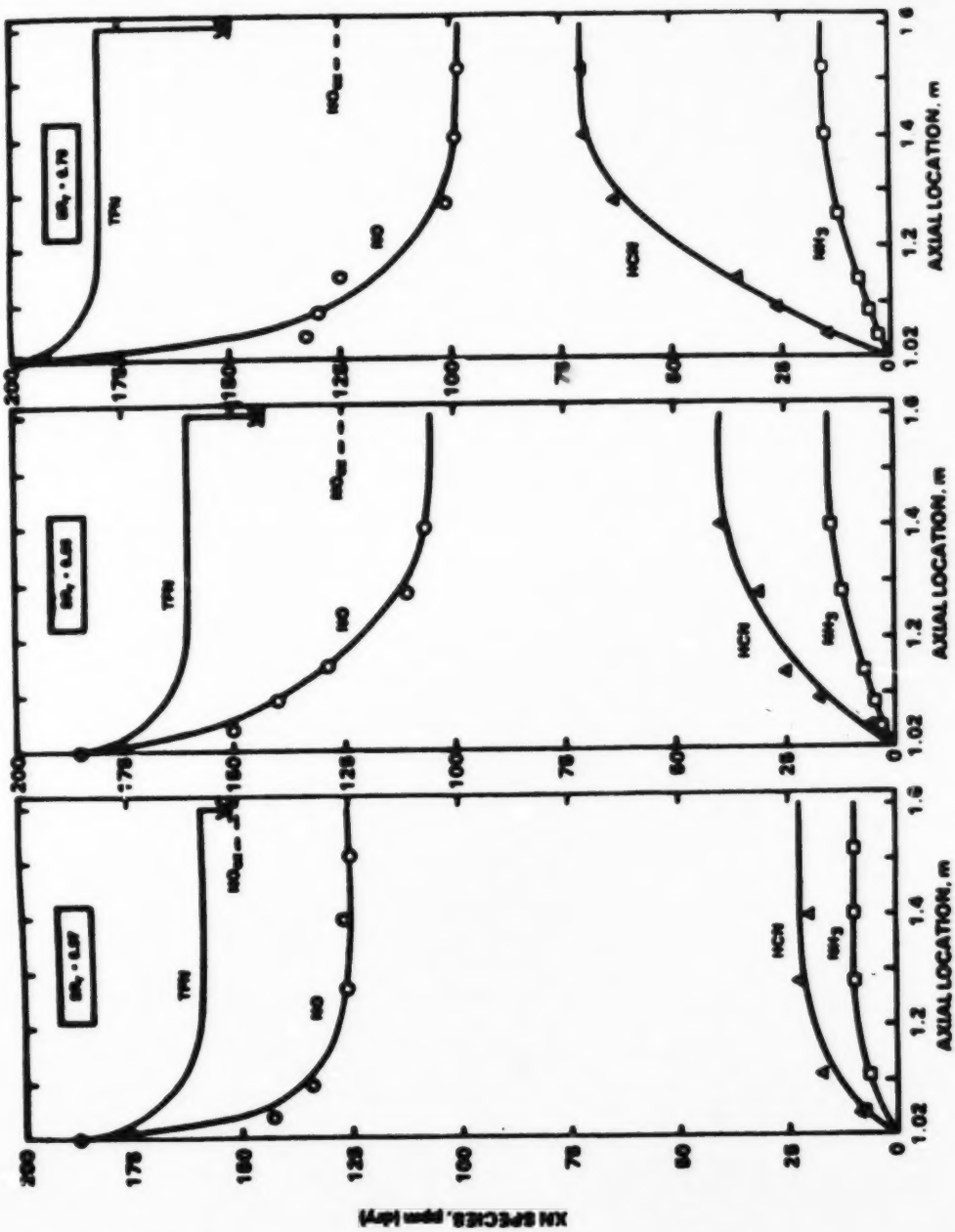

Figure 6. Fixed nitrogen species profiles in the reburn zone for NO_{pri} ~ 200 ppm.

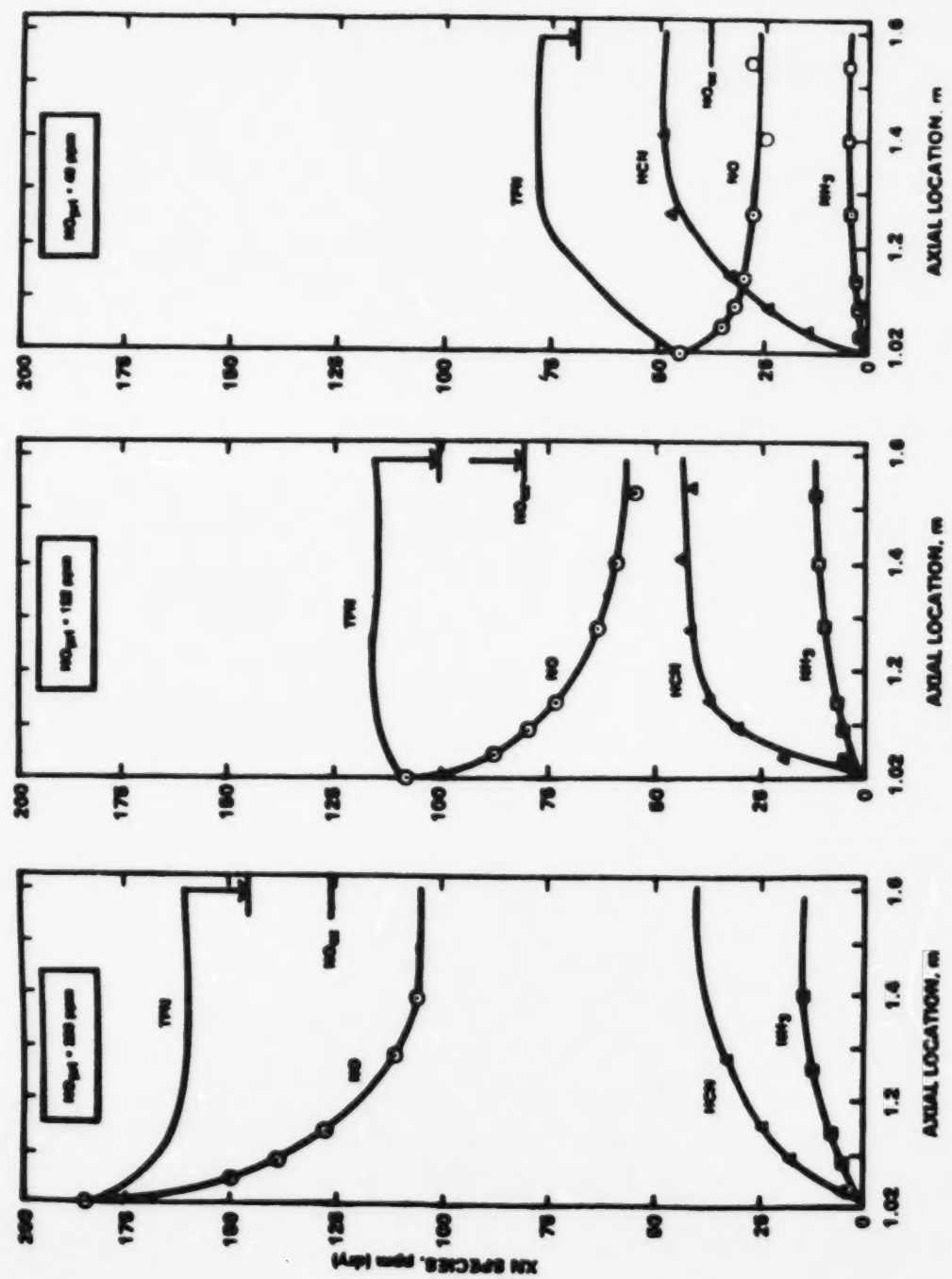

Figure 7. Fixed nitrogen species profiles in the reburn zone for $SR_r = 0.86$.

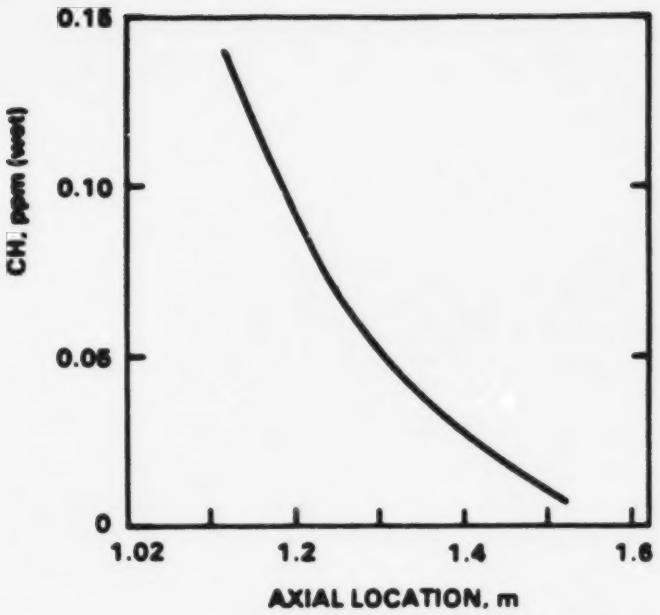

Figure 8. CH profile for $SR_r = 0.86$. This profile is calculated from Eq. (4).

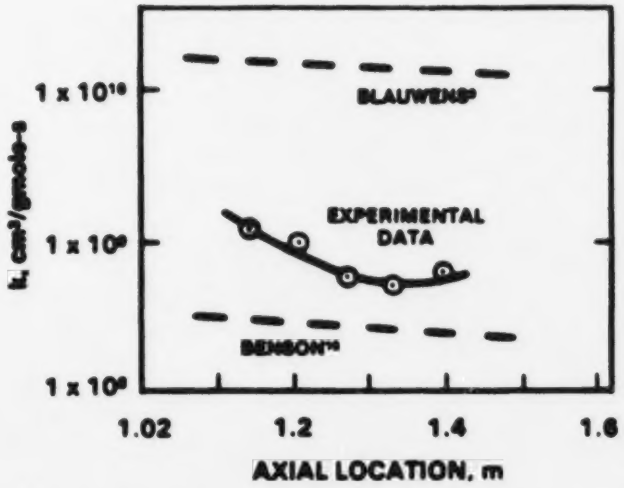

Figure 9. Rate constant for reaction 3. The experimental rate constant, calculated from Eq. (5), is compared with the rate constants reported by Benson, which are plotted as a function of reburn zone temperature.